

OAK RIDGE NATIONAL LABORATORY

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OAK RIDGE, TENNESSEE 37830

OFFICE OF THE DIRECTOR

October 3, 1977

Dr. James L. Liverman
Acting Assistant Secretary for
Environment
Department of Energy
Washington, D.C. 20545

Dear Dr. Liverman:

Enclosed is a copy of a report by Chester R. Richmond covering his recent foreign travel to Madrid, Spain, and Harwell and Windscale, England.

Sincerely,

A handwritten signature in dark ink, appearing to read "Herman Postma".

Herman Postma
Director

HP:br

Enclosure

cc: J. A. Lenhard-DOE/ORO
C. R. Richmond
File - RC

A handwritten signature in the bottom right corner of the page, possibly reading "H. Postma".

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ORNL

FOREIGN TRIP REPORT

ORNL/FTR-293

DATE: October 3, 1977

SUBJECT: Report of Foreign Travel of Chester R. Richmond, Associate Director for Biomedical and Environmental Sciences, Central Management Offices

TO: Herman Postma

FROM: Chester R. Richmond

PURPOSE: To visit key staff of the Junta de Energia Nuclear in Madrid, Spain, the National Radiological Protection Board and the Medical Research Council in Harwell, England, and the Chief Medical Officer of British Nuclear Fuels, Ltd., Windscale, England, to discuss health, safety, and environmental research and standards.

SITES VISITED:	9/7-9/10/1977	Junta de Energia Nuclear	Madrid, Spain	E. Iranzo
	9/13-14/1977	National Radiological Protection Board	Harwell, England	G. Dolphin
		Medical Research Council	Harwell, England	J. Vennart
	9/15-16/1977	British Nuclear Fuels, Ltd.	Windscale, England	G. Schofield

ABSTRACT: The traveler's discussions with Spanish and English personnel centered on health, safety and environmental aspects of nuclear materials, particularly the actinide elements. The traveler was primarily interested in activities related to man-related R&D and organizations concerned with radiation protection guidance and recommendations. Discussions with Junta de Energia Nuclear personnel concerned the accidental dispersal of plutonium at Palomares in 1966. Considerable information has been obtained since 1966, and it is possible that detailed reports for publication could be prepared early in 1978. It still appears that little, if any, plutonium has been incorporated into the residents of Palomares as the result of this accident.

The biomedical programs at the National Radiological Protection Board in England are excellent, and it is obvious that the English have selected important areas for study after reviewing the R&D in progress in the United States. Emphasis is now placed on establishing a life-time medical registry for radiation workers. Environmental and health problems discussed at Windscale were related to those encountered in an operating fuel reprocessing facility. The personnel visited at Windscale were also heavily involved with the national hearings on reprocessing which have been in progress since June 14, 1977.

VISIT TO JUNTA DE ENERGIA NUCLEAR, MADRID, SPAIN

Environmental contamination from plutonium in places such as Rocky Flats, Colorado; Mound Laboratory in Ohio; Thule, Greenland; Palomares, Spain; the Irish Sea; and the Bikini and Eniwetok Atolls of the Pacific has generated much concern throughout the scientific community. In addition to the need to learn more about these specific cases of environmental contamination, it is possible that we may deduce information that may be more helpful in the more general sense as regards environmental contamination arising from plutonium released from the nuclear fuel cycle associated with electrical power production or satellites using plutonium as a heat source in thermoelectric generators. The incident at Palomares, Spain in 1966 which resulted from an accident involving nuclear weapons created considerable international attention as it involved plutonium contamination of the general public in a foreign country. Since the accident on January 17, 1966, there has been interaction between the United States Atomic Energy Commission and subsequently the Energy Research and Development Administration and the Spanish Government, which has involved visits to Spain by members of the AEC/ERDA staff and contractor personnel, especially from the Los Alamos Scientific Laboratory and the Lawrence Livermore Laboratory.

The initial interaction between the United States and Spain resulted in an agreement between the United States and the Spanish Atomic Energy Commission which set up a four-point follow-up program designed to collect additional information on the Palomares accident. This program was to be conducted under the direction of the Junta de Energia Nuclear (JEN), Division of Protection and Medicine (under guidance of Dr. Eduardo Ramos), with equipment, technical help, and operational support supplied by the U.S. Atomic Energy Commission (Energy Research and Development Administration). The four-point follow-up program included the following:

1. Collection of information on the uptake and retention of plutonium and uranium by representative members of the population group who were potentially exposed to plutonium oxide by inhalation;
2. Measurement of the temporal and seasonal fluctuations in the plutonium air concentrations above the plutonium oxide-contaminated agricultural area that had been subjected to the decontamination procedures following the accident;
3. Serial measurements of contamination levels (both by plant uptake from the soil and by wind dispersal) of agricultural products produced in the contaminated area subsequent to decontamination; and
4. Studies of the temporal migration and redistribution of plutonium oxide in soil, decontaminated by deep plowing as a result of continued cultivation and weathering processes.

The traveler has been familiar with this particular contamination event (Project Indalo) since 1966 and has developed close working relationships with the key JEN staff, especially Dr. Iranzo who currently is in charge of the project.

Technical assistance amounting to approximately \$250,000 in the form of equipment, which included a whole-body and a chest counter, plus annual operating funds to the extent of approximately \$25,000 per year, has been provided by the U.S. Atomic Energy Commission (Energy Research and Development Administration) to further support the follow-up effort. In the fall of 1971, Dr. W. H. Langham of the Los Alamos Scientific Laboratory traveled to Palomares and Madrid to visit the scene of the accident some five and a half years later and to hold technical discussions with Drs. Eduardo Ramos and Emilio Iranzo and other members of the Junta's technical staff. Dr. Langham's report contained the following recommended actions to the AEC:

1. Encourage the Junta to publish their observations and data related to accident;
2. Increase operational support to the Junta and provide more technical staff;
3. Update the equipment used by the Junta and provide at least one additional alpha spectrometer;
4. Reevaluate their approaches to the four points of the agreement and modify them as indicated by the experience and information accumulated since the time of the accident;
5. Consider the advisability of providing the Junta with a lung counter to recount some of the Palomares residents who were examined during the first year following the accident; and
6. Manifest more interest in the work of the Junta through more review of their efforts.

Dr. Langham's recommendations resulted in a renewed interest on the part of the AEC in obtaining more information on the Palomares accident. Unfortunately, Dr. Langham was killed in an aircraft accident in May 1972, less than three months after this report was written. In early June, Dr. Ramos in a letter to Dr. H. D. Bruner of the AEC outlined some of the more pressing equipment needs of the Junta for use in Project Indalo. This ultimately led to the involvement of the traveler and that of others at the LASL in assisting the Junta via AEC Headquarters in updating equipment used at Palomares and Madrid in revitalizing the research and development program associated with Project Indalo. During FY 1974, the AEC/DBER made available \$27,000 capital equipment funds to be used for capital equipment for Project Indalo. It was LASL's responsibility to obtain the necessary equipment and, if necessary, to assemble and test the equipment prior to sending it to the Junta in Madrid. The

traveler's trip to Spain in 1974 was to visit Palomares and Madrid and to assist the scientific personnel at the Junta in developing future plans for Project Indalo. Subsequently, the equipment was shipped to Madrid and Mr. P. N. Dean of the LASL Health Division (now at Livermore) journeyed to Madrid in June 1974 to oversee the installation of the purchased equipment at the Junta in Madrid. Supervision was not provided for the installation of equipment to be used in the field at Palomares.

Since 1974, Phil Dean has visited the JEN on several occasions to provide advice on the chest counter (NaI - CsI) which is being used to monitor for the possible build-up of plutonium in people. The detector was not in use during the traveler's visit because additional shielding was being added to the lead lining of the steel counting chamber.

After the accident in 1966, 100 of the most likely exposed residents of Palomares were taken to Madrid and counted over the chest region by means of a proportional counter. The lower limit of detection was approximately 40 nanocuries, and no positive activities were determined in any of the measured individuals. The detector was later modified by the Spanish to give a minimum detectable limit of about 16 nanocuries, and several of the individuals who might have received higher exposures than others were counted. No positive measurements were observed even with the improved detector sensitivity. Forty-nine of the subjects were males over 14 years of age, 32 were females over 14 years of age, 10 were males under 14 years of age, and 9 were females less than 14 years of age. Measurements were made at the laboratories of the Division of Protection and Medicine of the JEN in Madrid. Prior to counting, the subjects underwent a complete medical examination in order to determine their state of health.

Attempts were made to search for plutonium in human subjects in 1975 and 1976. However, the data is all but useless because of calibration and other problems associated with the counting equipment (including high and variable background measurements). Also, only rarely have any of the subjects been measured more than once. Dr. Iranzo and this traveler decided on a follow-up study protocol which would involve annual measurements (chest counting, urine radiochemistry, medical examination, radiology, blood and urine chemistry, and chromosome analysis) on several groups of people. Others could be included should they express an interest. The subgroups would include the following:

- ° six people born since 1966;
- ° six people originally present in Palomares during the accident and nearest the highest contamination levels;
- ° six people working in areas currently associated with highest levels of residual contamination;
- ° six people who were not present in 1966;

- ° ten JEN staff who worked at Palomares after the accident; and
- ° six JEN staff who are not involved with Project Indalo.

Thus, about 40 people would be studied each year. Of these, 24 would be selected from the 1500 or so residents of Palomares. Six of the 40 people would have no connection with Palomares and would represent a control group.

It was also decided that it may be wasteful to spend too much effort on developing many lung phantoms for calibration purposes in view of the inherent biological limitations associated with chest counting.

The presently used techniques for chromosome analyses seem inadequate. The number of cells examined seem to be a function of the work load and currently only 50 cells are used. All new JEN employees provide blood samples for karyotyping and samples are routinely examined for uranium miners.

I mentioned that other organizations often scan a few cells to check for obvious genetically related syndromes and save the samples for reference and analysis at a later date should some question arise about exposure. It appears that more assistance is needed in this area.

Twenty-four-hour urine samples were collected from the same 100 individuals during their 1966-67 visit to Madrid. Three complete 24-hour urine samples were taken from each subject on three consecutive days. Alpha spectrometry was used to determine the plutonium-239 content of the urine. Seventy-one percent of the subjects showed no indication of urinary plutonium. Eighteen percent showed, in some of the analyses, plutonium contents of less than 0.1 disintegration per minute per 24-hour sample, and 9 percent showed activity levels between 0.2 and 0.1 disintegration per minute per 24-hour sample and 2 percent levels between 0.2 and 1.0 disintegration per minute in the 24-hour urine samples.

Additional samples were obtained from subjects who visited Madrid in 1975 and 1976. All the 1975 measurements were negative. Several of the 1976 measurements may have been positive and perhaps in the range of 0.05 pCi per 24-hours. Data for one subject suggest a value possibly 4 times higher.

For the planned study mentioned above, it is expected that a two-day urine sample (PM-AM/PM-AM) will be collected from each of the 40 subjects on a quarterly basis. Thus, up to 320 samples per year would need to be analyzed. Thus, additional analytical support (one person) and additional alpha spectrometers will be needed.

Considerable time was spent discussing the sampling program related to vegetation. Ashes from the sample preparation techniques are available for all samples collected from 1968 onward. However, ash samples are not available for samples collected in 1966 and 1967. Plutonium-239

measurements have been made from 1968 on, however, most every sample measured for the 1969 to 1972 period contains no plutonium-239. The main problem with data analysis is related to the occasional presence of plutonium particulates on the vegetation which gives spuriously high results. This effect is almost always noted in the wild vegetation and not in the cultivated crops. Many species of plants, both cultivated and wild, are measured and for the cultivated plants both foliage and fruit are measured for plutonium contamination. In virtually every instance the fruit of the cultivated crops show no plutonium contamination. Iranzo continues to obtain vegetation samples from outside the originally established study plots in an attempt to monitor for redistribution of the residual contamination. This will require more resources and will increase the number of samples to be measured. It might be useful at some future date to analyze some of the stored ash samples obtained from vegetation for Americium-241. This would require the acquisition of solid state measuring systems which would also be useful for soil analysis. Sampling Station 3-2 which was washed out in the 1973 flood has been reestablished.

Measurements of Airborne Alpha Activity. A meteorological station in the village needs attention and additional funds will probably need to be spent to upgrade the equipment. Unfortunately in 1969, two of the original four air sampling stations were discontinued and since then only two have been in constant use. It is quite interesting that the air samples measured during the past few years are all at the lower limit of detection, which is around 10^{-17} microcurie per cubic centimeter. Iranzo and colleagues are trying to establish possible trends in the change in air concentrations versus time since 1966. Mean plutonium-239 values for 1966 and 1967 were 0.28×10^{-15} and 0.09×10^{-15} microcurie per cubic centimeter, respectively. No particle-size measurements have been obtained to date.

It is unfortunate that the air sampling Station 2-1 was discontinued in 1969 since it was located in a position to sample possible pick up and movement of material from the untreated hillsides near Impact Point 2. Iranzo and I discussed the possibility of doing some selected air concentration measurements in the newly cultivated sections of Area 2 near Impact Point 2. I called attention to this problem in 1974, but unfortunately no action was taken.

Soil Measurements. During the last year or so Iranzo has concentrated on measuring plutonium in the upper soil surfaces (about a millimeter or so in depth) taken from the newly established sampling area designated 2.0. Thus, they are falling behind in their routine measurements associated with samples obtained from the five original sampling stations. It should be noted that for each of the five sampling stations measured per year, there are replicate measurements done for separate soil samples collected at numerous points on a grid for numerous depths resulting in 720 measurements per year ($8 \times 9 \times 5 \times 2$).

Plutonium-239 soil measurements were not started until 1969. A sample from which replicates are taken for analysis is stored for future

analysis. It will be possible to go back and measure these accumulated samples for Americium-241 should the occasion arise. There is currently a backlog of samples awaiting analysis. The current highest priority is on those samples taken from the soil surfaces and then the major priority will be to look at the depth profiles for samples collected in 1975, 1976 and 1977.

Iranzo and I agreed that it would probably be best to do the complete analyses for plutonium-239 at various depths at each location for 1969, 1972, and 1975, and to fill in the intermediate years at a later date. Hopefully, all the measurements for the upper soil surfaces will be completed by November of this year. I feel that it is imperative that additional equipment (alpha spectrometers) be made available for this project in view of the backlog of soil measurements and the large number of urine samples which will need to be measured in the near future.

I feel that it is extremely important to have a major publication on the "Palomares story" that will reach large segments of the scientific community and, in time, the general public. A considerable amount of information on the Palomares subject has appeared in print since 1966, but very often the reports are difficult to obtain. During my 1974 visit, I obtained several papers in Spanish and French which had been prepared by the JEN technical staff. The references attached to my 1974 report represent the bulk of the reports available on the subject.

The Hall-Otero agreement states that any publications should be jointly approved by the United States and Spain, and this constraint has been interpreted by some as one reason that a major publication has not been prepared. I do not believe this to be true. Dr. Iranzo and I discussed this topic in some detail, and I believe we agree on the need for a report in a widely read scientific journal.

Dr. Iranzo has agreed to visit ORNL next spring for the purpose of summarizing the available data for publication. I hope this can be accomplished, as the information would be useful to many organizations.

General Comments. I need to pursue many items related to my conversations with Iranzo and his staff. For example, techniques for the determination of small quantities of Americium-241, stabilization of the tracer used for analytical techniques for actinides, discussions with ERDA personnel concerning specific equipment needs, copies of certain ERDA contractor reports, techniques used at LASL for plutonium worker follow-up, EPA standards for drinking water and soils, liquid scintillation counting systems for plutonium, techniques used by industrial hygiene groups at several ERDA contractors, solid state equipment available for Americium-241 measurement, large volume air samplers, and information on particle size distribution of plutonium in soils.

VISIT TO NATIONAL RADIOLOGICAL PROTECTION BOARD

Major discussions with Dr. G. W. Dolphin centered on continued support from the United States (Oak Ridge National Laboratory) to Committee 2 of

the International Commission for Radiological Protection. We agreed that the ORNL will continue to provide active support in dose calculations for the ICRP and that this activity will not dissipate or be reduced. Dolphin was concerned that this activity might decrease because of the death of Dr. Walter Snyder. In fact, we both agree that the support of activities for dose calculations should be expanded as it is one way to get a larger commitment and greater participation from the United States into the ICRP. There has been mounting concern in the United States that the philosophy and thinking of the United States scientific community is not necessarily being reflected in policy actions being made by the ICRP. Dolphin has been helpful in obtaining better U.S. representation on the ICRP and its major committees. Some of our discussions in this area involved Dr. Jack Vennart who is the head of the Radiological Unit of the Medical Research Council which is also located at the Harwell site.

I spent a full day discussing research and development activities of the NRPB with Dolphin and his key staff members. It is most gratifying to see a first-class research program being developed by the English in the Biological and Medical area. The first report on the R&D activities of the NCRP published in 1976 reflects this young, but excellent, program. Dolphin is to be commended for his leadership in this area.

One of the most interesting efforts concerns the National Registry for Radiation Workers which was started on January 1, 1976. The object is to determine whether there are any differences in the causes of deaths and the ages of death of workers who had accumulated different radiation doses during their working lives and to assess the probabilities involved should any differences be found. First priority is being given to workers in the nuclear power industry, but it is planned to include other groups later on in the Registry. The names and other identifying details of the radiation workers will be recorded together with information on their exposures and doses. Each year information on subsequent exposures will be added and the records will be kept up-to-date in terms of changes in employment. Agreements have been made with the Office of Population Censuses and Surveys and with the General Registry Office of Scotland for the worker registry to be informed in time of the dates and causes of death for the participants. By agreement with those involved, the detailed information on the subjects will be strictly confidential and will be available only to the individuals concerned, their employer, and the National Radiological Protection Board. The system is quite straightforward in that once a name is entered into the Office of Population Censuses and Surveys by the NRPB, a response will automatically be made at some time in the future to the NRPB giving the date of death and the cause of death according to the internationally used coding system. This latter point is obviously an advantage in that it eliminates bias which could be introduced by various individuals within the NRPB assigning a cause of death in each instance. In a sense, the system is automatic once an entry is made and need only be updated periodically as regards input information. This activity is being led by John Reissland who is a physicist by training, but obviously has learned a great deal about epidemiological studies. There are about 70,000 workers in Britain who are exposed to ionizing radiations in the course of their work. The goal of this program is to include all radiation workers in Britain.

The NRPB is not part of the Atomic Energy Research Establishment (AERE) at Harwell as it functions under the Health Ministry with statutory requirements to conduct research and provide advice related to radioactive materials and electromagnetic radiations. The NRPB is conveniently located near the Medical Research Council which is also organizationally separate from the AERE at Harwell.

Major research efforts under Dolphin include the following: distribution and effects of actinide elements in rodents, including some interesting work on the use of plutonium-241 in autoradiographic studies; clearance of inhaled actinides from the lungs of experimental animals; deposition and clearance of inhaled aerosols in respiratory tracts; translocation of plutonium from simulated wound sites; solubility and chemical form of actinides in biological fluids; removal of incorporated radionuclides; actinides in the environment; the evaluation in dosimetry of radionuclides in man; radiation induced chromosome aberrations in human lymphocytes; population doses and risks; radiation physics in the National Registry for Radiation Workers.

I was interested to learn that small particles (1-2 nanometers diameter) of plutonium-dioxide rapidly solubilize if they are combined with sodium. This could have interesting effects as regards the combination of plutonium and sodium in breeder reactors. John Stather and colleagues are studying the accumulation of plutonium in the testes of man and animals as a follow-up to work which I reported several years ago. The data obtained from measurements of plutonium in tissues obtained at autopsy still suggest (as do data from the U.S.) that there is almost always less plutonium in the body as determined by autopsy, as compared to the estimate made by bioassay data during life.

Some of Dolphin's staff is involved in environmental measurements near the Windscale facility of the British Nuclear Fuels, Ltd. There is some concern that plutonium and americium that are released to the Irish Sea find their way back to the coast several miles south of the release point. They are taking measurements of plutonium in sediment samples along an estuary south of Windscale and are collecting air sampling data in an attempt to determine whether or not material becomes airborne from the sediments that are exposed to wind action during low tides. NRPB staff are also making beta-gamma measurements along the estuary south of Windscale. The NRPB also acts as a consultant to the Cumbria County Council in connection with BNFL's application for planning permission for an extension to its Windscale works. The NRPB is heavily involved with activities of the ICRP, the ICRU, the United Nations' Scientific Committee on the Effects of Atomic Radiation, the International Atomic Energy Agency, and the Nuclear Energy Agency (NEA) of the Organization for Economic Co-operation and Development (OECD).

I think there is a general need for increased interactions between R&D personnel in the United States and those of the NRPB.

BRITISH NUCLEAR FUELS, LTD., WINDSCALE

Dr. G. B. Schofield and I have exchanged technical information concerning the toxicity of actinide elements in man during the past several years and he has visited the United States on several occasions. Schofield is collaborating directly with the NRPB on the worker registry mentioned earlier in this report. Schofield arranged for me to tour most all the facility at BNFL except the exclusion areas related to plutonium. There are plans for extensive construction at the facility which could involve as much as three billion pounds. This includes new reprocessing facilities, new storage ponds for fuel elements, and areas for fixing waste products into glass matrices. In general, the general housekeeping and safety-related activities did not seem to be at the same level as those required in the USA. It is encouraging, however, to note that there are not a great many workers with plutonium burdens around the maximum allowable. I gathered on the average that there may be about one for each of the many years that the facility has been in operation.

While in the Windscale area, Schofield and I visited Whitehaven where a public inquiry was started on June 14 on the subject of nuclear fuel reprocessing. The inquiry was called by Peter Shore, Britain's Minister on the Environment. It has since been determined that a similar inquiry must be held on the subject of breeder reactors. Preparation for and participation in these hearings has been very stressful for many people at the BNFL as well as NRPB. Several well-known individuals from the United States have participated in these hearings in support of the Friends of the Earth which is leading the campaign against the BNFL in these hearings. It is expected that the hearings will continue for several more months. The inquiry is run by a commercial law judge, Mr. Justice Parker, who appears to be an extremely competent individual. These hearings should be of considerable interest to many organizations in the USA.

APPENDIX

ITINERARY

September 6, 1977	Travel from Oak Ridge, Tennessee, to Madrid, Spain
September 6-7, 1977	Project Indalo, Junta de Energia Nuclear, Madrid, Spain
September 12, 1977	Travel from Madrid, Spain, to London, England
September 13-14, 1977	National Radiological Protection Board and Medical Research Council, Harwell, England
September 15-16, 1977	British Nuclear Fuels, Ltd., Windscale, England
September 17, 1977	Travel from London, England, to Oak Ridge, Tennessee

PERSONS CONTACTED

Division of Protection and Medicine, Junta de Energia Nuclear, Madrid, Spain:

Dr. Emilio Iranzo, Head
Dr. Dolores Lara, Chemist
Dr. Francisco de Los Santos, Physicist
Dr. Celia Solas, Physician
Dr. Antonio Rebollar, Physician
Mr. Sinesio Salvador, Chemist

National Radiological Protection Board, Harwell, England:

Dr. G. W. Dolphin, Assistant Director, Research and Development
Dr. Hylton Smith, Head, Biology Department
Dr. John W. Stather, Applied Biology Section
Dr. D. C. Lloyd, Cytogenetics
Mr. N. D. Priest, Biologist
Dr. D. S. Popplewell, Actinide Biochemist
Mr. A. C. James, Aerosol Physics
Mr. J. Brightwell, Applied Biology
Dr. J. A. Reissland, Physics and Epidemiology

Medical Research Council, Harwell, England:

Dr. J. Vennart, Director, Radiobiology Unit at Harwell:

British Nuclear Fuels, Ltd., Windscale, England

Dr. G. B. Schofield, Chief Physician

LITERATURE ACQUIRED:

1. The Work of the National Radiological Protection Board 1974/1976. National Radiological Protection Board, March 1977, 73 pp.
2. NRPB/R&D 1. Annual Research and Development Report - 1976. National Radiological Protection Board, March 1977, 154 pp.
3. N. D. Priest, The Distribution of Pu-241 in Rodents. Int. J. Radiat. Biol. 31(1):59-78 (1977).
4. D. S. Popplewell et al., The Chemical Form of Plutonium in Urine. Radiat. Res. 62:513-519 (1975).